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Centro de Investigaciones  
Energéticas, Medioambientales  
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	DPTO. IMPACTO AMBIENTAL DE LA ENERGIA	
	PROTECCION	
	RADIOLOGICA POR INTERVENCION	
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Barret N. Fountos  
Program Manager  
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U.S. Department of Energy  
19901 Germantown Road  
Germantown, MD 20874-1290

Madrid, 20 July 2000

Dear Dr. Fountos:

Please find attached the following documents concerning the INDALO Project:

- Work Performed during 1999 (3 copies).
- Annual Work Proposal for 2000 (3 copies).
- CIEMAT forms for Cost Statement and Rates.
- Sheet of Cost Statement for the period 01-01-99 to 12-31-99.
- Report CIEMAT/DIAE/PPRI/51100/3/99 "Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear (Primer Semestre del Año 1999)".
- Report CIEMAT/DIAE/PPRI/51100/2/00 "Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear (Segundo Semestre del Año 1999)".
- Report CIEMAT/DIAE/552/55260/05/99 "Informe de la Estación Meteorológica de Palomares (Almería) 1998".
- Final Report. CEC FOURTH FRAMEWORK PROGRAMME. CONTRACT N° F14-CT95-0026-INHALATION OF RADIONUCLIDES, July 1999.
- J. Martínez, A. Espinosa, A. Aragón "Determinación de la variación en la concentración de partículas en aire a causa de las labores que implican movilización de suelos contaminados" 25 Reunión Anual de la Sociedad Nuclear Española. Granada (España. Nov. 1999.



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- A. Espinosa, A. Aragón, J. Martínez and J. Gutiérrez "Palomares Experience in Pu Environmental Behaviour". OIEA Contractor's meeting on the Coordinated Research Project on Site Characterization Techniques used in Environmental Restoration. Río de Janeiro (Brazil). April 1999.
- C. Gascó et al. "Participación del CIEMAT en estudios de radiología en ecosistemas marinos europeos". Publicación Técnica ENRESA. Nº 07/99.

If you need some more copies of the attached documents, please let me know.

Yours sincerely,

J. Gutiérrez  
Subdirector Gral. del Dpto. Impacto Ambiental de la Energía



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## **PROJECT INDALO**

### **WORK PERFORMED DURING 1999**

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#### **SUMMARY**

On September 15, 1997 a new agreement between the U.S. Department of Energy (DOE) and the Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT) was signed to establish a framework for scientific and technological cooperation by the Parties in radiological studies resulting from the accidental release of fissionable materials that occurred in Palomares, Spain, on January 17, 1966.

During 1999, the activities related to the environmental and personal radiological monitoring have been continued. Following the recommendations made by the expert panel in its 1998 report, special emphasis has been put on the soil studies concerning migration of Pu in zone 2.0 and on Pu analysis in zone 3.

The activities performed in 1999 are detailed in the two semi-annual reports sent to the Spanish national regulatory body, Consejo de Seguridad Nuclear (CSN). These two reports are attached. Also, as a result of the research carried out, papers have been presented to conferences and reports have been delivered in the framework of European Union Programme. These papers and reports are also attached.

Concerning the follow up of the measurements of soil contamination, a sampling of surface and deep soils has been carried out in zones 2.0 (8 core samples between 6 and 15 m. depth), 3 (121 surface samples) and 5 (15 surface samples). In total, 144 soil samples have been collected during 1999, although the 8 core samples will be divided in several sections producing a higher number of samples for analysis. In addition to these sampling activities, the following analyses have been performed: 216 analyses of Pu (radiochemistry followed by alpha spectrometry), 132 analyses of Am (gamma spectrometry) and 5 analyses of isotopic U (U-238, U-234 and U-235 by radiochemistry followed by alpha spectrometry).

The analyses of Pu are related to soil samples collected in 1998 in several areas of zone 2 and in zone 3. The following summary the obtained results:

- The results from zone 2 are different depending on the sampling area. In the NE side of area 2.1, that was affected by the original contamination plume and further cleaned by removal of the top 10 centimeters, a very heterogeneous range from less than 0.2 Bq/K (LLD) to slightly more than 2,000 Bq/K was found. The range for the side of area 2-1 closer to area 2.0, but not so affected by the plume, was from less than 0.12 Bq/K (LLD) to 36.3 Bq/K. The LLD (Lower limit of detection) slightly varies from sample to sample depending on parameters of the radiochemical separation and measurement processes, such as the chemical yield, the time of measurement, etc. These results correspond to soil surface samples.

- In zone 3, higher Pu values were found, also for soil surface samples. So a range from 3 Bq/K to 48,600 Bq/K was obtained. The higher values correspond to a uncultivated area, closer to impact point 3. For the cultivated area in zone 3 values around 100 times lower were obtained.
- In area 2.0, the results of two core samples (up to 50 cm deep) showed that the contamination remains in the top 5 centimeters (more than 90%). However the total amount of Pu found in the two cores was very different (4,781 KBq/K and 27.7 KBq/K), confirming the high values and the distribution previously observed.

The analyses of Am are also related to samples collected in 1998 in zones 2 and 3. The following summary describes the obtained results:

- As in the case of Pu, the Am values are very dependent on the sampled area. So, in the NE side of area 2.1, 14 of 54 samples showed values above the LLD, the highest value was of 1,990 Bq/K. In the side of area 2.1 closer to area 2.0, but not so affected by the plume, only 1 sample from 32 showed a positive value of 5.3 Bq/K, the rest of them being below the LLD. These results correspond to soil surface samples.
- The results from zone 3 showed higher Am values in a very heterogeneous range from 2.7 Bq/K to 11,000 Bq/K. As in the case of Pu, the higher values correspond to the uncultivated area close to impact point 3. For the cultivated area in zone 3 values around 100 times lower were obtained. These results also correspond to soil surface samples.

According to the provisions for work in 1999 the isotopic ratio Pu-239+240 / Am-241 has been obtained from data of 45 samples of surface soils collected in previous years, all of them with activity values for both radionuclides much higher than the LLD. In summary, the activity of both radionuclides shows a good linear correlation, the correlation coefficient being better than 87%. The estimated activity ratio was around 6. These data have been used for the calculation of Pu-241 activity in the bombs in 1966 and to estimate, from this value, when the highest activity of Am will be reached. The results obtained can be seen in a related document attached.

The analyses of isotopic U were performed in the core sample of area 2.0 with higher values of Pu. The results showed total amounts of U-234, U-235 and U-238 of 707 Bq/K, 28.3 Bq/K and 154.8 Bq/K respectively. The distribution with depth varies for the three isotopes and only from the section between 20 cm and 50 cm can the relation corresponding to natural U be observed. The different composition compared to that of natural U in the top 20 cm could be explained by the presence of U in the bombs.

In relation to the air monitoring, the sampling of air particles at stations 2-0, 2-1, 2-2 and P have continued, changing the filter on a weekly basis. The air volume collected is about 10,000 m<sup>3</sup> per sample in average. However, station 2-1 has been out of service from 09.04.99 until 21.05.99 and from 23.06.99 until the end of the year due to several problems in the power supply and obsolescence of the equipment (this equipment has been replaced for a new one in April 2000). Also station P was stopped until the end of June 99 due to a public reclamation based on excessive noise. Station P restarted at this date but stopped again on October

29, 1999 because problems in the air aspiration pump. These problems were repaired and station P was re-started in March 2000. In total 131 air samples were collected during 1999. The samples from 1998 have been analyzed and measured for Pu-239+240 and Am-241. Samples from station P were already analyzed and measured for Pu-239+240 during 1998 and showed Pu values lower than 1  $\mu\text{Bq}/\text{m}^3$  except for March 98 (14.5  $\mu\text{Bq}/\text{m}^3$ ) and July 98 (1.2  $\mu\text{Bq}/\text{m}^3$ ). Values of Pu ranged, during 1998, from 0.1 to 12.6  $\mu\text{Bq}/\text{m}^3$  in station 2-0, from 0.4 to 6.2  $\mu\text{Bq}/\text{m}^3$  in station 2-2 and from 0.2 to 30.8  $\mu\text{Bq}/\text{m}^3$  in station 2-1. Values of Am did not exceed 1  $\mu\text{Bq}/\text{m}^3$ , except during March in station P (3.9  $\mu\text{Bq}/\text{m}^3$ ) and during September and October in station 2-0 (1.6 and 1.2  $\mu\text{Bq}/\text{m}^3$ ). In addition to the mentioned Pu and Am air analyses, 4 samples were randomly chosen to determine the isotopic U content. U-238 values ranged from 3.0 to 5.1  $\mu\text{Bq}/\text{m}^3$ , U-234 values ranged from 3.3 to 5.7  $\mu\text{Bq}/\text{m}^3$  and values for U-235 were always around 0.2  $\mu\text{Bq}/\text{m}^3$ . In all cases, the relation among the values obtained fit well with the natural U composition. In total, around 90 analyses have been performed during 1999.

For vegetables, 18 samples of different crops (watermelons, wheat, tomatoes, etc.), have been collected in 1999. These samples were further divided in different parts (leaves, edible, etc.) and analyzed separately in order to know the type of contamination (external or internal) and its distribution. Analyses of Pu-239+240 in 28 samples collected during 1998 and 1999 have been carried out, corresponding to a total of 60 analyses due to the above reason. The results obtained for the edible parts of the samples were generally low, in the order of few tens of mBq/K. Only for a few samples higher values were detected (about 1 Bq/K in a olive fruit, 0.7 Bq/K in a tomato sample and 0.4 Bq/K in a wheat grain sample). Higher Pu values were detected for the external parts of some of the plants, but the value of 7 Bq/K was not exceeded, remaining, in general, lower than 1 Bq/K. In relation to Am-241, a total of 117 measurements by gamma spectrometry, corresponding to 61 samples collected during the years 1993 and 1995, have been carried out. The results were below of the detection limit in most cases; only in a few cases were positive values detected; these values always were detected in the leaves of the plants.

Concerning milk, 4 samples were collected, analyzed and measured for Pu-239+240; three of them were cow's milk and one goat's milk. Only two cow's milk had a positive result of 2.4 and 3.4 mBq/l; the other two remained below the detection limit.

Duplicate analyses of 5 snail samples collected in 1997 were performed. They were divided in shell and meat samples for analysis. In total, 18 analyses for Pu and also 18 analyses for Am were carried out. The obtained values differ largely. For the shell samples, the range of Pu values was from lower than the detection limit to 220 Bq/K. For the meat samples, the Pu values ranged from 0.3 to 901 Bq/K. Concerning Am values, the range for shell samples was from lower than the detection limit to 47 Bq/K and the range for the meat samples varied from 0.1 to 204 Bq/K. In all cases the lower values correspond to the sample collected close to air station P and the higher values correspond to the sample collected in the 2.0 area.

Due to the scarce rains during 1998 the rain water samples were not suitable for analysis (a very small volume was collected).

During 1999, 150 people from Palomares have been transported to CIEMAT headquarters in Madrid for medical examinations and sampling of 24-hour urine collections and further bioassay analyses and internal dosimetric assessments. In relation to Pu, the measurements of the 35 urine remaining samples from the 148 people examined during 1998 have been completed. Only two of them resulted in positive values: 1.19 and 0.50 mBq in urine of 24 hours, corresponding to one woman and one man respectively. All those whose urine samples tested positive values are requested to return to Madrid during the following year to confirm the existence and magnitude of their internal contamination. However, since the examinations are performed on those who agree to participate, not all those requested for follow-up examinations are re-evaluated during the following year. Some individuals choose not participate for different (personal) reasons. The measurements of the 150 people examined during 1999 have also been completed for Pu. Only four of them resulted in positive values: 1.14, 1.20, 0.70 and 0.41 mBq in urine collected over 24 hours. Those with positive values are different from those with positive values in 1998, one of whom was re-examined in 1999 with negative (lower than LLD) value. The rest of those with positive values in 1998 were also requested for follow-up in 1999 but they declined to travel to Madrid. Concerning Am, the urine analyses of the 150 people controlled during 1999 have been carried out. From these, 140 measurements have been completed, being all the results obtained below the detection limit of 0.37 mBq in urine of 24 hours. The remaining 10 samples are being measured at present. During 1999 no measurements in the whole body counter were made. The individual results of bioassay for each of the persons who are annually examined, are registered and sent to each of them, jointly with the result of their medical examination. These individual data are confidential. No significant findings related to radiation exposure were reported concerning the medical examinations (150 people) performed during 1999.

In relation to the marine ecosystem, the area accumulating most of the land-to-sea transferred transuranics has been located in the adjacent continental shelf, in the Aguas river submarine canyon. The sediments containing  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  are retained by the southern wall of the mentioned canyon, blocking their further transport southwards. Two new cores collected at the slope of the main canyon have been processed, about 60 radiochemical analyses have been performed. The isotopic ratios showed extra contributions of Pu and Am arising from the Palomares accident, confirming that this submarine channel is an active passage for contaminated sediments and associated radionuclides from the continental shelf to deeper areas. The final fate of the transuranics deposited onto the sea-floor has also been evaluated, since a number of environments where sediments act as radionuclides sources have been described. The geochemical association of plutonium and americium has been performed, showing that fallout Pu is mainly attached to the organic matter (humic and fulvic acids) and sesquioxides (Fe, Al and Mn). Fallout americium is majorly associated to carbonates (due to its similarity to calcium size and chemical behavior) and organic matter. Transuranics originating from the accident prevail in the residual fraction (i.e., largely irreversibly held), suggesting that they appear as refractory

compounds, likely as very fine oxide particles. Transuranics are linked to very stable phases, with low chemical reactivity (organic matter, clays, refractory particles) indicating that their post-depositional mobility is restricted to physical dispersion of the sedimentary material they are attached to, caused by turbidity currents, slumping phenomena, bioturbation, etc.

Concerning biokinetic studies of  $^{237}\text{Neptunium}$  oxide (Americium's daughter) the results showed that in spite of  $^{237}\text{NpO}_2$  being considered by ICRP to be a type M compound, a re-classification of the type of solubility of this compound could be suggested. However, further research on this topic would be necessary in order to confirm this conclusion. The final report for the European Commission Programme, partially supporting this research, has been produced and it is joined as an attached document.

These two last types of activities, marine and biokinetic studies, were conducted until June 1999, also partially supported by the European Commission, into the 4<sup>th</sup> R&D Framework Program and, in a lower extension, by ENRESA (Spanish National Company for the Management of Radioactive Wastes).

Finally, a report containing all the 1998 meteorological information collected by the CIEMAT's station located at the site has been produced. The report describes the characteristics of the meteorological stations and the analysis, interpretation and graphical representation of the data obtained. This report is also attached.

### **ATTACHED DOCUMENTS**

- Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear. (Primer Semestre del Año 1999). CIEMAT/DIAE/PPRI/51100/3/99
- Vigilancia Radiológica en la Zona de Palomares. Informe al Consejo de Seguridad Nuclear. (Segundo Semestre del Año 1999). CIEMAT/DIAE/PPRI/51100/2/00
- Informe de la Estación Meteorológica de Palomares (Almería) 1998. CIEMAT/DIAE/552/55260/05/99
- Final Report. CEC FOURTH FRAMEWORK PROGRAMME. CONTRACT N° F14-CT95-0026- INHALATION OF RADIONUCLIDES. July 1999
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